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ShieldingRequirements for the Transport of Nuclear Warhead Components Under Decommissioning

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SHIELDING REQUIREMENTS FOR THE TRANSPORT OF NUCLEAR WARHEAD COMPONENTS UNDER DECOMMISSIONING

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ABSTRACT

The requirements to carry out accurate shielding calculations involved with the safe off-site transportation of packages containing nuclear warhead components, special assemblies and radioactive materials are discussed. The need for a) detailed information on the geometry and material composition of the packaging and radioactive load, b) accurate representation of the differential energy spectra (dN/dE) for the neutron and gamma spectra emitted by the radioactive materials enclosed in the packaging, c) well-tested neutron and photon cross section libraries, d) and accurate three-dimensional Monte Carlo transport codes are illustrated. A brief discussion of the need for reliable dose measurements is presented.

1.0 INTRODUCTION

International agreements at the end of the cold war have resulted in the decommissioning of some of the nuclear weapons presently existing in storage facilities in the US and Russia. The transport of nuclear weapons components to disassembly plants and storage facilities must comply with stringent regulations to ensure the protection of the public health and safety and the environment.

In the US, the Department of Energy [1] (DOE) is responsible for assuring the safe packaging and offsite transportation of nuclear explosives, nuclear weapon and explosive components, special assemblies (i.e., weapon-like) and radioactive material. DOE requires that a safety analysis report for packaging (SARP) be prepared before the shipment of any special radioactive assembly. The SARP must document the structural, thermal, containment, radiation shielding, criticality safety, safe operating procedures and associated maintenance and fabrication acceptance criteria to guarantee a safe and reproducible packaging design. The SARP must show quantitatively to the members of the DOE Safety Transport Panel that the packaging and its radioactive contents comply with or exceed the requirements stated in the Code of Federal Regulations (CFR), Title 10, Part 71(10 CFR 71)[2] and Title 49, Parts 170 to 189 (49 CFR 170-189)[3] and other Code sections.

DOE has issued a design guide[4] on each of the areas that must be documented in the SARP as a guidance for its preparation by the applicant facility. However, alternate methods of design are equally valid once they are proved to comply with the CFR requirements.

In the present paper I will discuss the shielding issues that must be considered in order to carry out an accurate calculation of the photon and neutron dose rates at the outside surface of the package. Together with the accuracy of the calculations, the magnitude of the dose rates must comply with certain allowed upper limits imposed by government regulations. These limits determine the requirements on the packaging to assure that it provides the appropriate shielding for the radioactive content.

For clarification purposes the terms packaging and package in this paper are defined as follows: **Packaging** means the assembly of components necessary to ensure compliance with the maximum dose rates allowed outside the package. The packaging may consist of one or more receptacles, absorbent materials, spacing structures, thermal insulation, radiation shielding, and devices for cooling or absorbing mechanical shocks. The special vehicle to transport the package and the tie down system for the package inside the vehicle may be also designated as part of the packaging. **Package** refers to the packaging together with its radioactive content as prepared for transport.

2.0 SHIELDING DESIGN CONSIDERATIONS

The shielding design considerations for the packaging of special radioactive materials are dependent on the method of shipment, the dose rates expected as a result of the package contents and its form and conditions of transport. The conditions of transport can be those expected as normally experienced in the shipment of a package (normal conditions of transport, NCT), or unusually severe conditions as a result of an accident (hypothetical accident conditions of transport, HACT) which potentially could jeopardize the function of the packaging by preventing the shielding of the radiation from the load, or resulting in the loss of criticality control from fissile contents.

Accordingly, the packaging must perform the following functions under NCT and HACT: 1) containment of the radioactive material, 2) shielding to minimize the radiation escaping the package and 3) prevention of criticality, and 4) in addition, the packaging must protect the safety function (thermal and structural) components present in the package, such as the cooling system, thermal insulation, pressure vents and impact limiters.

2.1) The containment system in the package consists of the holding vessel with the associated gaskets and a closure. The integrity of the containment must be demonstrated by a series of structural and thermal tests as specified by government regulations.

2.2) The shielding provided by the packaging must comprise gamma ray shielding and neutron shielding. The gamma ray shielding designed to attenuate the photon dose rate from the fission and decay gamma ray spectra high density is effectively obtained by high Z materials (steel, lead). Depleted uranium may also be used as a photon shielding material as long as the its neutron emission is shielded. The neutron shielding designed to attenuate the neutron dose rate from the fissile contents and (α,n) reactions is optimized by using Li or B loaded hydrocarbons which moderate and absorb the neutron flux.

2.3) The subcriticality of the fissile contents of a package can be assured in different ways. A standard method is to introduce "neutron poison" materials among the shielding materials that will capture the neutrons before they can cause further fissioning of the contents. Another method is to adjust the geometry of the radioactive load to facilitate the neutron leakage from the package and from arrays of packages. In this case, however, the neutron dose rate outside the package must not exceed that allowed by shielding regulations.

2.4a) The function of the thermal safety component is to give thermal protection to the package from external heat sources, as well as from the heat load generated by the radioactive decay of the contents. Insulation materials accomplish the protection from external heat, while conductors or surface enhancement techniques such as fins are used to remove the heat generated by the radioactive decays inside the package. The thermal design must solve the conflict of passively removing the radioactive decay heat while passively protecting the package shielding and containment structures from accidental external heat sources. For example, in the US Code of Federal Regulations (10 CFR 71.43 and 49 CFR 173.442) the criterion related to the thermal load of the package states that 1) the internal generated heat cannot affect the integrity of the package and 2) in still air at 38°C(100°F) and in the shade, no accessible surface of the package would have a temperature exceeding 50°C if the package is transported in a non-exclusive shipment.

For most packages the thermal insulation is constructed from wood products or other cellulosic materials or from other organic materials or foams. In these materials (wood, polyurethane foam, cellulose, cork) the main components are hydrogen, carbon and oxygen that make them good neutron shielding materials. However, these materials may decompose at elevated temperatures (as could exist in an accident during transportation) with the release of combustible gases that may ignite in the presence of air with possible coupling of the combustion energy to the package. Since existent regulations in the United States forbid the use of active cooling techniques, preferred materials will be those that no change of phase or decomposition will take place in the temperature range expected in NCT or HACT.

2.4b) Among the structural safety components one has pressure vents and impact limiters. The vents are used to relieve pressure buildup inside the package to prevent excessive mechanical stresses inside the package. However these vents must be designed such that the release of radioactivity does not exceed the maximum limits stated by the regulations.

2.4c) The function of the impact limiters is to reduce the effect on the shielding and containment vessels of dynamic loads acting on the external packaging. Materials that exhibit good mechanical energy absorption characteristics frequently also have good thermal insulation properties and work well as a neutron shielding. For these reasons, the impact limiter may also function as the thermal insulation to protect the containment vessel and the radiation shielding, together with enhancing the shielding.

3. SHIELDING CRITERIA FOR THE TRANSPORT OF RADIOACTIVE MATERIALS

In the US, federal regulations and/or recommendations by agencies including DOE, the Nuclear Regulatory Commission[5] (NRC) and the International Atomic Energy Agency[6] (IAEA) define the criteria for the packaging and shipment of radioactive materials. Some of the regulations deal specifically with shielding issues in the transport of radioactive packages under normal and hypothetical accident conditions of transport. The objective of these regulations is to protect the public health and the safety of the environment from uncontrolled release of radioactive contents and loss of shielding from the packaging.

The shielding criteria for normal conditions of transport (found in 10 CFR 71.47 and 49 CFR 173.441) define the upper limit for the total dose rates (sum of photon and neutron dose rates) at any point on the external surface of the package and the transport index of the package. (49 CFR 172.400). The transport index is defined as the maximum value of the total dose rate in millirem (mrem) at 1 m of distance from the package surface.

The values for the dose rate upper limit and index of transport are determined by the conditions of transport. If the package is sharing the transport vehicle with other packages then the upper limit is set by these regulations equal to 200 mrem/hour with a transport index of 10. However, the conditions of transport maybe more stringent such as 1) the package is to be shipped in a closed vehicle, 2) the package position in the vehicle is fixed during transportation, and 3) no loading or unloading operations take place between the initial and final destination of the package. Under these conditions the dose rates at the surface of the package can be as high as 1000 mrem/hour while a value of 200 mrem/hour is the maximum allowed at any point on the external surface of the vehicle and 10 mrem/hour at any point 2 m from the surface of the conveyance.

The upper limit for the dose rates established by the CFR under hypothetical accident conditions of transport is 1000 mrem/hour at 1 m of distance from any point on the surface of the package. It is reasonable to assume that in a serious accident the surface of the package has suffered severe damage and the external container has been destroyed and the insulation material placed between the external and internal container has been has been burned. Under these conditions the regulations require a maximum allowed dose rate of 1000 mrem/hour at 1 m of distance from the surface of the inner container.

Furthermore, the allowed escape of radioactive materials from the damaged package must not exceed A_2 Curies in a week. The values of A_2 have been calculated for each radionuclide accordingly to its nature as gamma-ray, x-ray, β-ray, or α emitters. The A_2 values have been tabulated[2] for all known radionuclides in Appendix A of 10 CFR 71.

4. VALIDATION OF THE PACKAGING SHIELDING DESIGN

To show that the design of the packaging selected to transport a given radioactive load will comply with regulations stating the maximum dose rates allowed outside the package, the shielding design needs to be validated. Calculational methods are needed to validate the shielding design of the packaging before the actual container (packaging) is built. Undoubtedly, a good set of measurements of the dose rates at the surface of the package with the radioactive material in place is the most effective way to get the radiation levels outside the package and to validate the calculational method used to obtain these dose rates.

Federal regulations in the US require that calculation of the dose rates be done before the container design is approved for the transport of special radioactive materials or warhead components. The calculations must be done for normal and hypothetical accident conditions of transport. The dose rates must be calculated on the outside surface of the package at locations where it is expected that their magnitude is larger due to the positioning of the radioactive load inside the package.

Customarily, the dose rates are calculated at the top and bottom of the outside container and around the side surface at a height determined by the position of the load in the inside container. To obtain the "index of transport" of the package these calculations are repeated at a distance of 1 m from the outside surface of the package. For HACT, the calculation is done for the same positions chosen for the NCT, but in this case the dose rates will be calculated at 1 m of distance from the inside container. The accuracy of these calculations will be scrutinized by a DOE panel before the container is certified to transport a given load.

An accurate calculation of the photon and neutron dose rates requires the following points be included:

4.1 Package Calculational Model

One must start from a detailed model of the weapon parts or radioactive materials to be transported, the packaging system with the inner and outer containers, and the insulation layers. The three-dimensional model of the package components must follow very closely the mechanical drawings of the weapon system and packaging. The densities of the materials present in the package with their isotopic and chemical compositions must also be included in the calculations. This information will allow a first test of the calculational model by a comparison of the calculated volumes and masses of the load and packaging with those measured and given in the mechanical drawings. Attention must be paid to the location of the radioactive source (or sources) with respect to the external surfaces of the package and the existence of irregularities in the packaging that could result in streaming channels for neutrons and gammas.

4.2 Source Calculations

An accurate calculation of the gamma and neutron source intensities resulting from the radioactive materials in the package must include the isotopic composition, impurities and the age of the sources.

a) Isotopic Composition. In the calculation of the photon source the isotopic composition of the fissile material must include those small percentages of isotopes that are generally omitted, such us ^{232}U or ^{241}Pu. If enriched uranium is present in the package, all $^{232,234,236,238}U$ isotopes must be included. For example, the presence of ^{232}U (even in a most negligible quantity) accounts for the production of high energy photons <E> 2.0 MeV in the source spectrum, with the emission of 2.614 MeV gammas from ^{228}Tl (daughter product from an α-decay chain of ^{232}U). For a plutonium load, the $^{236,238,241,242}Pu$ isotopes must be included along with the $^{239,240}Pu$ isotopes, which are usually the only ones included in the source calculation. The presence of ^{236}Pu ($t_{1/2}$ = 2.851 years) will account for the emission of the 2.614 MeV photons through the ^{232}U-decay chain. The presence of ^{241}Pu (β-decay to ^{241}Am) with a half life of 423.7 years, will significantly increase the intensity below 100 KeV.

b) Impurities. The impurities in the radioactive materials (elements other than the actinides) are important in the calculation of the neutron source. Impurities found in weapon type materials range from Li to rare-earth metals and their presence results in an increased neutron flux due to the contribution of (α,n) reactions on the impurities. For example, the intensity of the neutron source from enriched uranium (94% ^{235}U) could increase as much as a factor of two with the presence of certain impurities.

c) Age of the Source. The age of the fissile material is an important parameter in the calculations, since the amounts of ^{228}Tl and ^{241}Am will increase with time in enriched uranium and plutonium loads respectively. For example, for enriched uranium the intensities of the neutron and photon source reach their peak at about 10.5 years; while for a Pu load the neutron source peaks at 10.5 years and the photon source at 70.5 years.

d) The specific code used to calculate the neutron and photon sources, after the information in a) to c) has been included, will determine the accuracy of the calculations. The ORIGEN-S[7] code does an accurate calculation of the neutron and gamma-ray spectra from radioactive materials by including fuel depletion, actinide transmutation, fission product buildup and decay and associated radiation terms. The neutron spectrum includes both the contribution of the fission neutrons and that from (α,n) reactions. When running the source calculations, care must be taken in choosing the energy interval structure for the gamma ray spectrum. The best criteria is to have the width of the energy intervals small enough to bracket the gamma peaks observed in the measured spectrum. Wider intervals could be chosen for the continuous regions of the spectrum. The neutron source can be run with coarser intervals given the smooth character of the neutron spectra from fission.

4.3 Neutron and Photon Transport Codes

The choice of a code to calculate the neutron and photon dose rates outside the package has to be based on the characteristics of the whole system (materials and geometry). The choice of one-, two-, or three-dimensional transport calculations will be dictated by the geometry of the entire package. In general one finds that the complexity of the package often requires the use of a three-dimensional MONTE CARLO transport code to do an accurate calculation of the dose rates. MCNP,[8] TART,[9] COG,[10] and MORSE[11] (in this order), are the most accurate and best known codes to be used in dose rate calculations. These codes are well documented.[12]

4.4 Neutron and Photon Cross Sections

The neutron and photon cross sections used in the above Monte Carlo transport codes are from the ENDF/B data system[13] or the ENDL[14] library. The cross sections in these libraries have been extensively compared with differential and integral measurements through the years. Cross section evaluation groups at Brookhaven, Los Alamos and Lawrence Livermore National Laboratories are continuously revising and updating the libraries when new measurements or evaluations become available. Version VI is the last revision of the ENDF/B library released by Brookhaven, although version V is used yet in many of the calculations such as those done with ORIGEN-S.

4.5 Conversion Factors from Fluxes to Dose Rates

The gamma and neutron fluxes calculated with the above codes will be obtained in units of flux; gammas/$cm^2 \cdot s$ and neutrons/$cm^2 \cdot s$. To convert the fluxes to dose rates (rem/hour or mrem/hour) conversion factors are needed. The American National Standard Institute (ANSI/ANS6.1.1) has tabulated[15] these factors as a function of photon (0.01 to 15 MeV) and neutron (2.5e-08 to 20 MeV) energies. In most of the codes, these factors can be entered in the input file to get the output directly in dose rate units.

5.0. MEASUREMENTS

Accurate measurements are always a valuable tool to test the calculational model assumptions and simplifications made to facilitate the calculations. A good set of measurements requires the use of the appropriate set of detectors to obtain the neutron and photon fluxes outside the package. Absolute calibration of these detectors must be done against standard gamma and neutron sources. The measurements could be done at the same positions as the calculated dose rates. For the comparison with the HACT calculations, the measurements can be done at the surface of the inner container before the whole assembly of the package is completed.

Discrepancies between measurements and calculations are frequent even if the codes and cross sections used in the calculations have been tested against benchmark measurements and the detectors are well calibrated. Even in the absence of other radioactive materials near the package, reflection from the ground or concrete floors, nearby walls and sky-shine effects could contribute to higher measured dose rates. In order to minimize the in-scattering into the detector, the package and the detector need to be suspended by a light frame in open space, several meters above the ground or nearby walls and massive equipment (such as large magnets).

6.0 SUMMARY

The shielding requirements involved in the transport of special nuclear materials and warhead components as a result of disassembly and storage operations have been discussed. In order to assure the safety of the public and environment, the government must establish regulations to determine the maximum values of the dose rates allowed outside a package carrying these radioactive loads. These upper limits will be different according to the type of conveyance and conditions of transport. Official mechanisms must be established to assure that laboratory's storage and disassembly sites have guidance to follow the regulations and comply with them. Well documented calculations of the dose rates previous to any shipment between sites are encouraged in order to validate the container design and shielding properties. Accurate calculations of the dose rates require a detailed model of the radioactive load and packaging system and accurate calculation of the neutron and gamma sources in the package. These calculations require input of all isotopes present in the actinide materials (even those present in minuscule quantities as is the case of $^{232,234,236}U$ or $^{236,238,241,242}Pu$) in order to have a good description of the source/sources gamma spectrum. Also input of all impurities present in the fissile materials (from Li to rare-earths) which will give rise to higher intensities of the neutron source due to the (α,n) reactions on the impurities. The age of the source is an important parameter in the calculations since the intensity of the neutrons and gammas are a function of the number of years the weapon components have been in storage. In enriched uranium components, the intensities of both neutron and gamma sources will peak at about 10.5 years, while for Pu components the gamma source peaks at around 70.5 years and the neutron at 10.5 years. The use of three-dimensional Monte Carlo codes for the transport of the neutrons and photons to the whole assembly and cross sections' libraries that have been tested against bench mark experiments. Measurements of the dose rates complement the calculations and they are a valuable tool in testing their accuracy.

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